

Bright source of $K\alpha$ and continuum X rays by heating Kr clusters using a femtosecond laser

R. ISSAC, J. WIRTHIG, E. BRUNETTI, G. VIEUX, B. ERSFELD, S.P. JAMISON, D. JONES,
R. BINGHAM, D. CLARK, AND D.A. JAROSZYNSKI

Department of Physics, University of Strathclyde, Glasgow, Scotland, United Kingdom

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Abstract

X rays emitted from Kr clusters illuminated by a femtosecond laser have been observed over a wide spectral region from 3 keV to 15 keV. The measured spectra are characterized by a broad bremsstrahlung continuum and $K\alpha, \beta$ lines at 12.66 keV and 14.1 keV. To the best of the authors' knowledge, this is the first observation of $K\alpha, \beta$ emission from laser-heated Kr clusters. The bremsstrahlung continuum arising from collisions in the plasma implies a population of hot electrons consistent with a temperature of several kiloelectron volts. The absolute X-ray yield in the 3–15 keV region is found to be of the order of 10^7 photons per laser pulse. The plasma temperature, estimated from the continuum part of the spectrum as a function of laser intensity and X-ray yield as a function of laser pulse duration, are studied.

Keywords: Cluster nanoplasma; Intense lasers; Rare gas clusters; X-ray generation

1. INTRODUCTION

The development of X-ray sources based on high-peak-power lasers has been one of the thrust areas of research since the advent of the table-top terawatt chirped pulse amplified (CPA) laser. This has stimulated the effort to understand processes involved in intense laser interaction with matter. Laser-produced plasmas from optically ionized gases, clusters, and solids have been widely explored as sources of X-ray and extreme UV over the last decade (McPherson *et al.*, 1993; Ditmire *et al.*, 1996; Giulietti & Gizzi, 1998; Rocca, 1999; Hulin *et al.*, 2000; Junkel-Vives *et al.*, 2001; Krainov & Smirnov, 2002). Solids and van der Waals clusters of rare gases have been found to have excellent X-ray yields and useful spectral content under intense laser interaction. Rare gas clusters have solidlike local densities, and therefore absorb laser radiation very effectively while at the same time producing negligible debris upon disintegration (Ditmire *et al.*, 1996). Laser absorption mechanisms in clusters and plasma hydrodynamics have been subject to intense study since the discovery of unprecedented X-ray flux from intense laser interaction with clusters (McPherson *et al.*, 1993). Monitoring high-energy electrons (Shao *et al.*, 1996;

Chen *et al.*, 2002), ions (Dobosz *et al.*, 1999; Nishihara *et al.*, 2001), neutrons (Zweiback *et al.*, 2000; Schwoerer *et al.*, 2001), and X rays (Ter-Avetisyan *et al.*, 2001; Skobelev *et al.*, 2002) has helped to unravel some of the processes underlying the intense X-ray emission. Initial investigations suggested that Mie resonance of the spherically shaped clusters and resonance heating at the critical density surface could explain most of the observations (Milchberg *et al.*, 2001). Further investigations where the laser pulse duration was varied found the duration to play a major role in determining particle (ion and electron) energy spectra and total X-ray flux (Zweiback *et al.*, 1999; Parra *et al.*, 2000; Kumarappan *et al.*, 2002). Plasma expansion and radiative blast waves, which occur long after the laser pulse transit, introduce further ionization in the plasma (Edwards *et al.*, 2001). X-ray spectra have helped to characterize these sources over the years, and so far the highest observed X-ray line energy from cluster targets has been identified as 3.3 keV, identified as K -shell emission from H-like argon (Junkel-Vives *et al.*, 2001). The presence of a population of hot electrons was found to be responsible for the excitation of line spectra (Abdallah *et al.*, 2001). However, these observations were made over a restricted spectral window and therefore spectral information over a much wider spectral band, particularly at higher energies, remained unknown. To date, all reports of X-ray emission from clusters have concentrated on X rays from ions with L - and M -shell vacancies

Address correspondence and reprint requests to: D.A. Jaroszynski, University of Strathclyde, Department of Physics, John Anderson Building, 107 Rottenrow, Glasgow ONG, Scotland, United Kingdom. E-mail: dino@phys.strath.ac.uk

from atoms of high atomic numbers (e.g. Kr, Xe) and $K\alpha$ lines of lighter atoms (e.g., Ar, Ne, N). In the work presented here, we have observed, for the first time, X-ray bremsstrahlung over a broad spectral region and intense $K\alpha$ radiation at 12.66 keV from krypton irradiated by a focused femto-second laser beam with an intensity of less than $3 \times 10^{16} \text{ Wcm}^{-2}$.

2. EXPERIMENTAL PROCEDURE

The experimental chamber used for the laser–cluster interaction experiments, shown in Figure 1, consists of an ultra-high vacuum six-way cross connected to a larger chamber that is equipped with a high-throughput turbomolecular pump that attains a base pressure of 10^{-8} torr and maintains 10^{-3} torr during experiments. Krypton gas with a stagnation pressure of 7 bars or 15 bars is pulsed at a repetition rate of 10 Hz through a cylindrical nozzle with a throat diameter of 2 mm to form clusters. The high-pressure supersonic gas jet forms a cloud of atom clusters held together by van der Waals forces when the gas is cooled by adiabatic expansion through the nozzle. Measurement of the cluster size distribution is usually difficult to carry out without partially destroying the clusters; therefore, Rayleigh scattering of 400-nm laser beam from the jet has been monitored to establish the formation of clusters. Theoretically, the Rayleigh scattered signal (S) should scale roughly as the cube of the backing pressure, $S \propto P_0^3$ (Ditmire *et al.*, 1996). However, in our own measurements, as in some other experiments (Ter-Avetisyan *et al.*, 2001), the signal of the scattered light scales with the square of the stagnation pressure, $S \propto P_0^2$. A semi-empirical parameter, called the Hagen parameter, determining the average cluster size for an axisymmetric cylindrical nozzle is $\Gamma^* = kd^{0.85}p_0/T_0^{2.29}$, where d is the nozzle throat diameter

in micrometers, p_0 the backing pressure in millibar, T_0 the initial gas temperature in degrees kelvin (K), and k ($= 2890$ for Kr) a constant that depends on the sublimation enthalpy per atom and van der Waals bond length of the gas (Wormer *et al.*, 1989). The cluster size scales as $N_c = 32(\Gamma^*/1000)^{2.25}$ for $\Gamma^* > 1000$ (Hagena, 1992). According to this scaling law at room temperature ($T_0 = 293$ K) and stagnation pressure 7 bar, we estimate the mean number of atoms per cluster as $\langle N_c \rangle \sim 6.5 \times 10^4$, which gives an estimate of the radius of clusters as $R_c \approx r_0 \langle N_c \rangle^{1/3} \sim 10$ nm and the corresponding quantities for 15 bar is 3.6×10^5 and ~ 17.5 nm, respectively. The density is $\sim 10^{23} \text{ cm}^{-3}$, of the order of solid density.

Part of the energy from a 5-TW CPA Ti:sapphire laser ($\lambda = 800$ nm, $\tau_p = 50$ fs) beam is used to irradiate the cluster cloud using a $f = 100$ mm UV-grade fused silica lens with a measured focal spot radius of $8 \mu\text{m}$. The laser pulse duration is adjusted by varying the grating separation in the pulse compressor of the CPA laser system. Single photon counting is used to measure X-ray spectra in the range 2–20 keV using a Peltier cooled Si:PIN detector, which gives a measured energy resolution of 350 eV at 14.4 keV and excellent detection efficiency in the spectral range. Single X-ray events are accumulated using a gated multichannel analyzer calibrated with a Co^{57} source. The spectra are corrected for the efficiency of the entire detection and measurement system (e.g., filter and window transmission) prior to analysis (X-ray transmission data tables obtained from www.cxro.lbl.gov). The count rate is kept below 0.1/pulse to avoid distortion of the spectra due to pile up by limiting the detection solid angle to less than 5×10^{-4} sr using apertures.

3. RESULTS AND DISCUSSION

$K\alpha$ and $K\beta$ emission from krypton at 12.66 keV and 14.1 keV, respectively, has been observed, as shown in Figure 2, for a

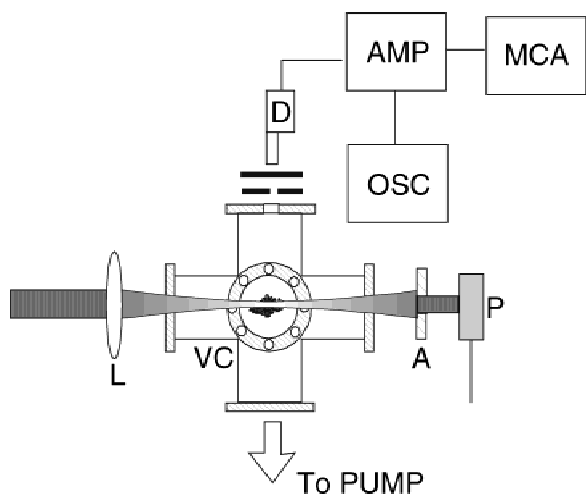


Fig. 1. Experimental system used for X-ray observations from clusters. L: focusing lens; P: Laser power meter; D: X-ray detector (XR-100CR, Amptek); AMP: spectroscopic amplifier; MCA: Multichannel analyzer; OSC: Oscilloscope; VC: vacuum chamber; A: variable aperture.

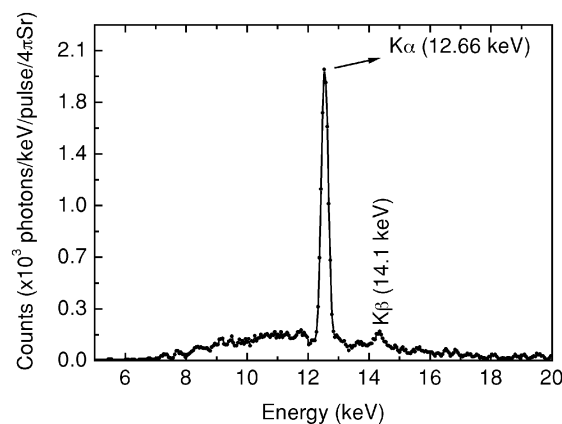


Fig. 2. X-ray spectrum transmitted through 350- μm aluminum filter showing the K-shell radiations from Kr cluster plasma. Integrated X-ray yield across $K\alpha$ line profile after correcting for filter transmission is $\sim 10^5$ photons/pulse/ $4\pi\text{sr}$.

cluster radius 10 nm and a laser intensity of $2 \times 10^{16} \text{ W cm}^{-2}$. The laser intensity is an order of magnitude lower than that used in many similar experiments (Dobosz *et al.*, 1997) with substantially larger clusters. The low energy continuum part of the spectrum is filtered using a 350- μm -thick aluminum filter to isolate the K -shell radiation having $\sim 28\%$ transmission at 12.66 keV. The measured X-ray spectra corrected for filter transmission factors, shown in Figure 3 as a function of laser intensity, have broad features corresponding to a bremsstrahlung continuum in the 3–15 keV region. These spectra are qualitatively similar to those found in condensed phase experiments (Korn *et al.*, 2002). The strong resemblance of the spectra to that emitted by solid density matter and the fact that unclustered gas targets cannot be heated efficiently to extreme high temperatures indicate that absorption and emission happens while the high density phase is sustained. After the clusters have expanded and form uniform plasma, the density has fallen drastically so that collisional effects are minimal. The continuum emission originates from either free–free or free–bound transitions during interactions of electrons with ions and can be effectively used as a diagnostic of the temperature of the plasma (Yaakobi *et al.*, 1996). For both free–free and free–bound transitions, the spectrum depends exponentially on photon energy, as $I(h\nu) \propto \exp(-h\nu/k_B T_e)$, which indicates a Maxwellian distribution of electrons, where h is Planck’s constant, k_B the Boltzmann constant, and T_e the electron temperature. Thus the slope of the semi-logarithmic plot directly gives the electron temperature of the plasma. The thick solid lines in Figure 3 are exponential fits to the measured spectra. A minimum of $6.8 \times 10^{15} \text{ W cm}^{-2}$ is required to produce measurable $K\alpha$ radiation. Furthermore, the line to continuum ratio grows as a function of laser intensity due to more effective heating to higher temperatures. The onset of $K\alpha$ radiation indicates that hollow Kr atoms with K -shell vacancies are being formed by collisions with high-energy electrons. Since it is not possible that these inner

shell electron excitations responsible for $K\alpha$, β emissions are produced through direct field ionization by the relatively low laser intensities used here, the dominant mechanism responsible must be excitation due to high-energy inelastic electron–ion collisions. For Maxwellian electron distributions, the cross section for electron impact ionisation is (Brunner, 1997)

$$\sigma = 4.5 \times 10^{-14} n_j \frac{g(\eta/\eta_j)}{\eta_j^2},$$

where n_j is the number of electrons in the shell j , η_j is the ionization energy in electron volts, η is the kinetic energy of the electrons, and $g(\eta/\eta_j)$ has the form

$$g(\eta/\eta_j) = \eta_j \frac{\ln(\eta/\eta_j)}{\eta}.$$

From the above, the required electron energy to enable inner shell vacancies must be equal to or greater than the ionization energy, which for the krypton $1s$ electron is 14.3 keV. Observation of $K\alpha$, β lines is therefore evidence of the existence of electrons with energies higher than 14.3 keV. The electron temperature evaluated from the continuum spectrum is shown in Figure 4. The solid line is a least squares fit of the data to $kT_e \propto I^{1/2}$, which is more slowly varying than the ponderomotive energy scaling, $U_p = 9.33 \times 10^{-14} I$ (W cm^{-2}), which rules out predominance of direct ponderomotive heating mechanism.

The high local density within clusters, which is of the order of solid density, and the low average density of the medium, demands that the initial absorption and heating occurs within the cluster prior to expansion. Even though anomalous flux of X rays from clusters was observed almost a decade ago, none of the existing models fully explains laser–cluster interactions. Ditmire *et al.* (1996) proposed a nanoplasma model that still remains the most successful model for predicting anomalous heating of clusters. In this

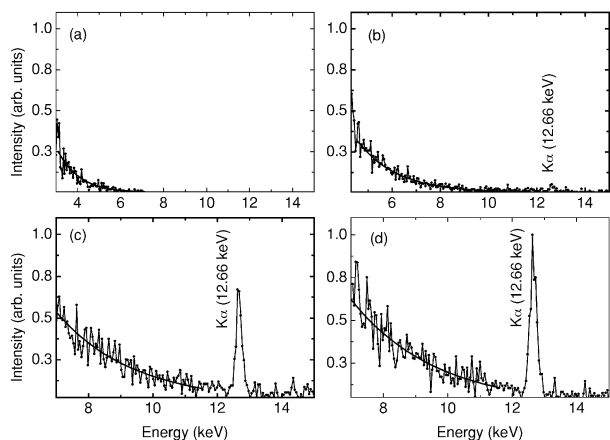


Fig. 3. X-ray emission from $R_c = 4$ nm krypton clusters irradiated with a laser of intensity (a) 2.5×10^{15} (b) 6.8×10^{15} , (c) 1.2×10^{16} , and (d) $2 \times 10^{16} \text{ W cm}^{-2}$. Spectra are corrected for filter transmissions.

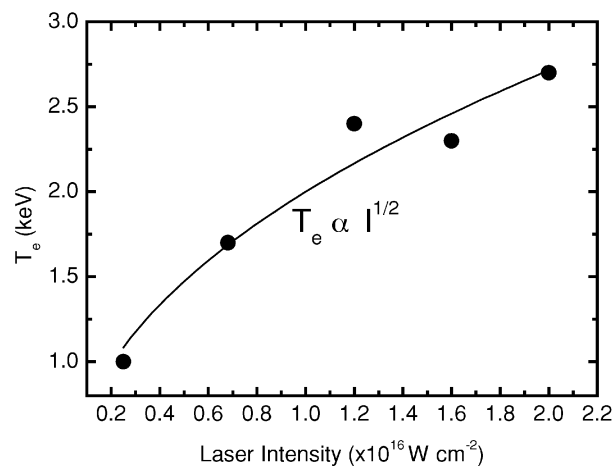


Fig. 4. Plasma temperature deduced from the continuum spectra measured as a function of laser intensity.

model, a nanoplasma is formed by rapid heating of the cluster by collisional processes followed by hydrodynamic expansion. The heating process encounters a strong resonance when the electron density is equal to $3n_{cr}$, where n_{cr} is defined for the excitation wavelength, $n_{cr} = \epsilon_0 m_e \omega^2 / e^2$, where m_e and e are the electron rest mass and charge, respectively, and ω is the angular frequency of the laser. The enhanced absorption happens because of the resonance in the dipole moment of the plasma sphere of radius R_c given by $\mathbf{P} = R_c^3 [(\epsilon - 1)/(\epsilon + 2)] \mathbf{E}$ where ϵ is the plasma dielectric function, $\epsilon = 1 - \omega_p^2 / [\omega(\omega + i\nu_{ei}/2)]$, ν_{ei} is the electron-ion collision frequency, $\omega_p^2 = n_e e^2 / \epsilon_0 m_e$ defines the plasma frequency, n_e , the electron density, and \mathbf{E} represents the laser electric field. \mathbf{P} undergoes resonance when $\epsilon + 2 = 0$ or $\omega_p^2 = 3\omega^2$ while $\omega \gg \nu_{ei}$. However, such a resonance has a short lifetime and can happen during the buildup and decay of the electron density according to simulations (Milchberg *et al.*, 2001). It has also been suggested that, in addition to the $3n_{cr}$ resonance heating, resonance absorption at the critical density surface will contribute to heating for large clusters (Milchberg *et al.*, 2001). Even though for clusters with dimensions much smaller than the wavelength of light defining the angle of incidence at poles of the plasma sphere is questionable and therefore resonance absorption at the critical density surface resembling laser–solid interactions is not fully justifiable when the electric field inside the cluster is uniform. Rhodes and colleagues (McPherson *et al.* 1993) formulated a model for small clusters that relies primarily on coherent intracluster collisions. The basic assumption of this model is that individual atoms inside the cluster are ionized through direct field ionization. It successfully explains the threshold for the appearance of L -, M -, and N -shell emission from xenon for clusters of $N_c < 10^3$ atoms. However, our observations of harder X rays at low laser intensities suggest that this scaling may not directly apply for K -shell emissions.

X-ray yield from 3 to 15 keV as a function of pulse duration for larger clusters ($R_c \sim 17.5$ nm) is measured and given in Figure 5. Total X-ray yield is more than 10^7 photons/pulse/ 4π sr. Quantitatively X-ray yield in our measurements is significantly higher than that observed for krypton clusters of comparable dimension and for ~ 20 times the laser intensity (Dobosz *et al.* 1997). The highest observed X-ray energy from krypton by Dobosz *et al.* (1997) was 1.75 keV, and any indication of K -shell emission at higher X-ray energies was absent. Figure 5 shows that there exists an optimum pulse duration at which the X-ray yield is maximum. For fixed cluster size and pulse energy, the integrated X-ray yield, which is a measure of absorption and electron–ion collisions, first increases significantly with increasing pulse duration toward a maximum at 300 fs and decreases for longer pulses. Because the laser energy per pulse is kept constant, at the highest peak intensity (shortest pulse duration) the X-ray yield is approximately 2.5 times smaller than that at the optimum pulse duration. The broad peak of the curve is between about 250 and 800 fs, which correspond to

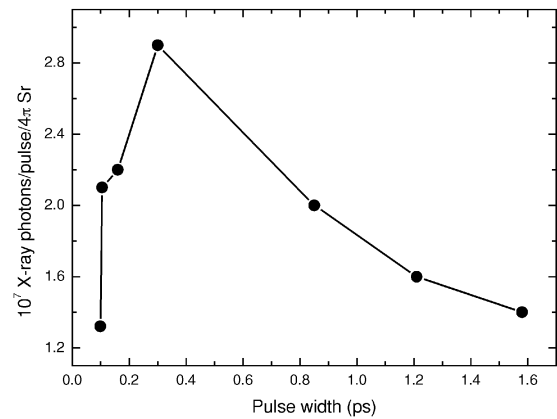


Fig. 5. Dependence of total X-ray yield for krypton clusters of radii 10 nm in the 3–15-keV region, as a function of laser pulse duration at fixed pulse energy. Laser intensity corresponding to pulse duration 250 fs is 2.4×10^{16} Wcm $^{-2}$.

an intensity of $(3\text{--}0.93) \times 10^{16}$ Wcm $^{-2}$. The electron oscillation amplitude in the laser field of magnitude E_0 at this intensity is $x_0 = eE_0/(m\omega^2) \sim 14\text{--}8.5$ nm (neglecting the dielectric response of the cluster). This is comparable to the cluster dimensions within predicted cluster size distribution $N_n \sim R_n \exp(-R_n^2/2R_c^2)$ (Parks *et al.*, 2001; Krainov & Smirnov, 2002).

In almost all models developed so far on cluster heating there is a general agreement that a substantial contribution originates from electron–ion inverse bremsstrahlung, though various resonances enhance the absorption when the conditions are right. During collisions, part of the energy can also be radiated as X-ray bremsstrahlung. When the oscillation amplitude is comparable to the cluster dimensions, there is significant overlap between the electron and ion clouds to facilitate effective collisional absorption. For the shortest pulses used here, the oscillation amplitude is ~ 25 nm and hence the electron cloud overlap with the ions is minimal during the laser pulse. Moreover, the ponderomotive energy for the electrons, U_p , is higher than the measured thermal energy with short pulses, and therefore the collision frequency scales with laser electric field as (Silin, 1965), $\nu_{ei} = Z n_e m_e \omega^3 \ln \Lambda / r \pi \epsilon_0^2 E_0^3$, and hence rapidly decreases with increasing laser field amplitude. Here $\ln \Lambda$ is the Coulomb logarithm. This reduction in collision frequency in conjunction with the larger oscillation amplitude contributes to less effective heating and X-ray yield with shorter pulses. For longer pulses than the optimum, the hydrodynamic expansion of the cluster reduces the density and thus the number of collisions. The efficiency of laser coupling to the cluster nanoplasma can therefore be enhanced by altering the electron dynamics in the laser field using variable pulse width irradiation as indicated by our results.

4. CONCLUSION

We have measured X-ray spectra in the region of 3 to 15 keV from laser-heated krypton clusters. The direct observation

of broadband bremsstrahlung radiation demonstrates the role of collisions during cluster heating and evolution. Dependence of X-ray yield on pulse duration is arguably an indication of collisional effects during laser cluster interaction through better overlap between electron cloud and the ion background in a single ionizing cluster. For longer pulse width (a few hundred femtoseconds) and moderate intensities, the oscillating electrons spend most of their time inside the cluster, enabling more energy to be absorbed through collisions with ions due to relatively small amplitude oscillations over a larger number of laser cycles. In the case of shorter pulses with same laser energy density, electrons are driven with higher oscillation amplitude and hence result in less effective electron–ion overlap and therefore less efficient collisional heating. A reduced total X-ray flux is observed for pulses shorter than 300 fs. Therefore, the results shows that careful selection of laser and cluster parameters enhances the X-ray spectral yield even at moderate laser intensities. Detailed and quantitative theoretical investigations to establish the effect of collisions in cluster heating are underway and will be published elsewhere in the near future.

Hard X-ray K -shell emission lines can potentially be used as a powerful source of ultrashort pulses of incoherent X-ray radiation for time-resolved X-ray diffraction and imaging applications in ultraclean environments. For large clusters, $R_c \sim 17.5$ nm, we have measured X-ray photon yield $> 10^7$ /pulse, useful for imaging, diffraction, and medical applications. Laboratory X-ray sources in the 10–20-keV region are particularly useful for medical applications like mammography. The required laser intensities are readily available in medium-scale laboratories and, furthermore, the source is debris free and can be operated at a reasonably high repetition frequency. Previous observations under similar experimental conditions have been limited to line emission with energies of a few kiloelectron volts, seriously limiting the applicability of the source.

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